

**Great Lakes Binational Toxics Strategy:  
The Level I Pesticides in the Binational Strategy**

**Prepared by  
Battelle**

**for  
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## **THE LEVEL I PESTICIDES IN THE BINATIONAL STRATEGY**

### **EXECUTIVE SUMMARY**

The Binational Toxics Strategy (BNS) identified twelve bioaccumulative substances having sufficient toxicity and presence in water, sediments and/or aquatic biota of the Great Lakes system to warrant concerted action to eliminate their input to the Great Lakes. They are called “Level I substances.” Six of the substances are formerly used pesticides, and are the primary focus of the two governments’ commitments related to pesticides. The Level I pesticides are aldrin, dieldrin, chlordane, DDT (plus metabolites DDE and DDD), mirex, and toxaphene. The BNS documents combine aldrin and dieldrin because aldrin is readily oxidized to dieldrin, and is rarely found in the environment. These Level I pesticides are covered by the following “Challenge,” written in the BNS:

*Confirm by 1998 that there is no longer use or release from sources that enter the Great Lakes Basin of five bioaccumulative pesticides (chlordane, aldrin/dieldrin, DDT, mirex, and toxaphene), and of the industrial byproduct/contaminant octachlorostyrene. If ongoing, long-range sources of these substances from outside of the U.S. are confirmed, work within international frameworks to reduce or phase out releases of these substances.*

Level I pesticides are the subject of this report, which was first issued as a “Draft for Public Comment” on December 31, 1998. Octachlorostyrene is covered under a separate report. The Level I pesticides are highly chlorinated compounds, with five or more chlorine atoms per molecule. They are bioaccumulative, and concentrate in fish and piscivorous birds, having been found to produce several negative effects on birds, including impaired reproduction due to egg shell thinning. They all have been shown to be probable carcinogens based upon laboratory studies with animals.

### **Historical Usage**

The past usage of these pesticides was large enough to cause significant environmental contamination during the years of their use. DDT, the first large scale pesticide, reached peak annual usage of some 80-85 million Kg in 1962. Toxaphene use peaked in 1972-75 at close to 30 million Kg per year. Other estimated peak annual use rates were chlordane at 12 million Kg in 1971, aldrin plus dieldrin at 9 million Kg in 1966, and mirex at 300-400,000 Kg in 1963-68. Again, the use rates of aldrin and dieldrin are combined because of the conversion of aldrin to dieldrin in the environment. The pesticide uses were the only significant application for the Level I pesticides with the exception of mirex. About 25% of the mirex production was for pesticidal uses, the balance being used as a flame retardant.

Because of the negative environmental effects of these substances, the pesticide uses of all of the Level I pesticides have been canceled for domestic use in the U.S. The flame retardant uses of mirex were curtailed in the 1970's and replaced by other products. All but chlordane have not been in production in the U.S. for many years. Chlordane continued to be produced in the U.S. for export by the product's sole manufacturer, Velsicol Corporation. In 1997 Velsicol announced that the production of both chlordane and heptachlor would cease. Velsicol expected to complete the shipment of existing stocks from its Memphis, Tennessee plant by the end of 1997.

### **Trends in Environmental Loadings**

While domestic production has ceased and pesticide uses have been canceled, these pesticides continue to have an environmental presence. That is not surprising, considering the large use rates of the 1960's and '70's coupled with their persistence and atmospheric deposition from long range sources. These pesticides continue to be produced and used in other countries, contributing to the atmospheric deposition. The environmental concentrations, however, have shown a general decline in most media over the years, with a few exceptions.

**Surface Water.** It is estimated that 22,474 Kg of Level I pesticides remain in the Great Lakes water as calculated from the most recent water concentration data (1994 - 1997):

aldrin + dieldrin	4,163 Kg	chlordane	308 Kg
DDT + metabolites	417 Kg	mirex	110 Kg
toxaphene	17,476 Kg		

All of these levels represent reductions over time with the exception of toxaphene. Lake Superior accounts for about 77% of the toxaphene calculated to be in the water of all five of the Great Lakes. The current water concentration level in Lake Superior can not be expected to change rapidly for several reasons; past inputs have remained in the lake because the low lake water temperature reduces vaporization loss, the low particulate volume in the lake water reduces the removals to sediments, and losses through outflow are small relative to the large lake volume (191 years average water residence time). Estimates calculated from the Pearson, Swackhamer data indicate that when net atmospheric inputs fall to zero (that is just equaling the vaporization loss) it would require over 40 years to reduce the toxaphene concentrations by one half.

**Sediment.** In general, the sediment core data are limited, and do not cover all substances in all lakes. Most cores analyzed for the Level I pesticides show the expected pattern of rising concentrations from the time of introduction to the peak use years, followed by declining concentrations thereafter. A few cores showed exceptions to this pattern. A recent analysis of toxaphene in sediment cores (Pearson, Swackhamer, et al, 1997) showed the expected concentration of toxaphene rising to a peak in the 1970-80 period, followed by a continued decline. These scientists concluded that atmospheric input is currently the dominant source of toxaphene to the Great Lakes, with the exception of Northern Lake

Michigan, which, they noted, may have a non-atmospheric source. In 1997, in search of the non-atmospheric source, a number of tributaries were sampled at locations that were felt most promising to elucidate the elevated toxaphene concentrations based upon past pesticide use and current industrial activity. Although final data have not been published, preliminary information indicates that non-atmospheric sources of toxaphene were not found. Another anomalous finding involved two of five Lake Michigan cores analyzed for chlordane, DDT and dieldrin which showed rising dieldrin concentrations in recent years. However, one of these, from the northern part of the Lake is inconclusive, as the chlordane and DDT peaks came in at about the year 1900, long before the commercial introduction of the pesticides. The other core from the southern part of the Lake needs confirmation, as DDT concentrations are 10 times those for chlordane and dieldrin.

**Atmosphere.** Environmental concentrations of the Level I pesticides in the Great Lakes Basin are affected by atmospheric transport. Atmospheric concentrations around the Great Lakes are being measured by the Integrated Atmospheric Deposition Network (IADN). Concentration data have been taken at the five master stations covering the Great Lakes. Time trend data are not available for all substances, but measurements for dieldrin, DDT and its metabolites, and three principal components of commercial chlordane are available for 4 to 5 year periods from 1990 through 1995. The data were corrected for temperature, and subjected to regression analysis; decreasing concentration trends over time were calculated for all of these compounds. Using the data that were significant at the 95% confidence level, rate constants were calculated and used to estimate the time required for the atmospheric concentration to reach the detection limit of  $0.1 \text{ pg/m}^3$ . The detection limit is one way to define “Virtual Elimination.” Using this definition, the estimates of future dates to reach virtual elimination ranged from about 2010 for DDT to about 2060 for the DDT metabolite DDE, with dieldrin and chlordane falling in between. Aside from the overall decreasing trend, unusually high seasonal atmospheric levels of DDT and its metabolites were measured near South Haven, MI. This area is presently under study in attempt to elucidate the reasons for the elevated concentrations, which might include vaporization from soils of past DDT use, inadvertent releases, or the present use of the pesticide dicofol, which contains DDT as a contaminant.

**Bioaccumulation.** Level I pesticides are still present in the tissues of fish and birds in the Great Lakes Basin. However, concentrations in fish and herring gull eggs have shown an overall decline over the years. An example is the reduction in the concentration of DDT in Lake Michigan lake trout from about 20 ppm to 1 ppm over the period 1970 to 1992. An exception again, however, is the concentration of toxaphene in Lake Superior lake trout, which showed no significant change from 1982 to 1992; this is most likely a result the higher and stable concentrations of toxaphene in Lake Superior water.

While environmental concentrations in the Great Lakes Basin media have been generally declining for the past twenty years, and current contamination levels are well below drinking water standards, concerns remain because the substances persist and bio-accumulate in fish and wildlife. There continue

to be fish consumption advisories based on unacceptable levels of these pesticides in sport and commercial fish.

### **Reservoirs and Unused Stocks**

There are over 100 National Priority Level Superfund sites within the eight Great Lakes States which show contamination by one or more of the six pesticides. A former Velsicol Chemical production site in St. Louis, Michigan, now under remediation, has considerable DDT contamination, and carp taken from the adjacent Pine River have high levels of DDT. These sites represent point sources that are being addressed through the U.S. EPA Superfund Program. In spite of these point sources, evidence of significant contaminant introduction to the Great Lakes beyond site boundaries has not been confirmed.

Overall removals of Level I pesticides at waste pesticide collections (so called Clean Sweeps) have resulted in significant recoveries of unused stocks. A simple illustration of their significance is the fact that the quantities collected have exceeded the total quantities in the Lake waters, and the quantities of chlordane, aldrin/dieldrin, and DDT collected are many times those levels. Although mirex has not been identified in Clean Sweeps, some mirex may have been collected in New York, as those collections identified all organo chlorines as DDT, and all cyclodienes as chlordane.

### **Options and Opportunities for Further Reductions**

The declining concentration trends for most of these substances is encouraging, and shows progress over the years. The current concentrations levels are well below drinking water standards, but one or more of the Level I pesticides are the subject of fish consumption advisories in each of the Great Lakes. Further declines are likely to be gradual, as net atmospheric and other inputs are balanced by removals by sedimentation and flow.

The processes available for further reductions are in place and on-going. These are:

1. Remediation of sites with contaminated soils and sediments under the Superfund Program. Clean-ups at a former DDT manufacturing site in St. Louis, Michigan and toxaphene contamination at a former manufacturing site in Georgia are examples of on-going work.
2. Waste pesticide collections by the States to continue the removal of stored stocks.
3. National efforts (PBT Initiative) to reduce emissions that can deposit in the Great Lakes.
4. International efforts (POPs Initiative) to reduce long range atmospheric transport.
5. Continued support for monitoring (IADN) to followed trends and investigations of anomalous situations to add new insights (the South Haven, MI study is expected to provide information about releases to the atmosphere from soils treated recently and in the past).

### **Has The U.S. Met The Challenge?**

All pesticide uses for all Level I pesticides have been canceled. The production facilities within the U.S. have been closed. Although evidence of purposeful releases has not been identified, the potential release from contaminated sites and remaining stockpiles is still possible. However, the declining concentrations indicate that such possible releases are likely to be small. Because some Level I pesticide concentrations in the Great Lakes are still above Water Quality Criteria, and fish advisories are required, continued monitoring is necessary. However, these options are in place and on-going.

For these reasons, we believe that the United States has met the principal intent of the Challenge, even though the statement “...no longer use or release...” can never be confirmed as long as unused stocks and contaminated sites exist.